Stratospheric chemistry and aerosols

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Stratospheric chemistry

Gas phase chemistry

1. Chapman Cycle
2. Catalytic cycles
   1. Hydrogen radicals (HO_x)

$OH + O \quad H + O_2$
$H + O_3 \quad OH + O_2$
Net: $O + O_3 \quad 2\,O_2$

$OH + O \quad H + O_2$
$H + O_2 + M \quad HO_2 + M$
$HO_2 + O \quad OH + O_2$
Net: $O + O_3 \quad O_2$

$HO_2 + O \quad OH + O_2$
$OH + O_3 \quad HO_2 + O_2$
Net: $O + O_3 \quad 2\,O_2$

Hydrogen Source Gases: H_2O, CH_4

- Long term trends
- HO_x chemistry in the upper stratosphere and mesosphere

Stratospheric chemistry

2. Nitrogen radicals (NO_y)

$NO_2 + O \quad NO + O_2$
$NO + O_3 \quad NO_2 + O_2$
Net: $O + O_3 \quad 2\,O_2$

Nitrogen Source Gas: N_2O (and ...)

- Long term trends
- NO_y partitioning (in the lower stratosphere: aerosols)

3. Chlorine radicals (Cl_y)

$ClO + O \quad Cl + O_2$
$Cl + O_3 \quad ClO + O_2$
Net: $O + O_3 \quad 2\,O_2$

Chlorine Source Gases: Organic Chlorine

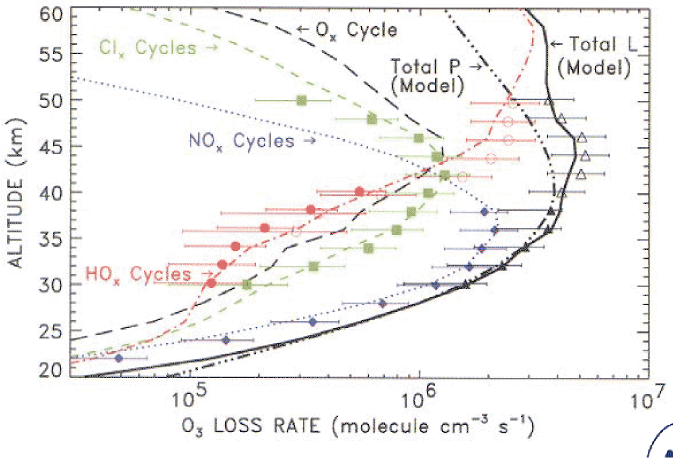
- Long term trends
- Cl_y partitioning (in the lower stratosphere: aerosols)

Ozone loss

35°N,September 1993

24 hour average

WMO, 1998



Lower Stratosphere

Gas phase chemistry triggered by heterogeneous processing

ClO + ClO + M → ClOOCl + M

ClOOCl + h → 2 Cl + O_2

2 (Cl + O_3 → ClO + O_2)

Net: 2 O_3 → 3 O_2

BrO + ClO → Br + Cl + O_2

→ BrCl + O_2

BrCl + h → Br + Cl

Br + O_3 → BrO + O_2

Cl + O_3 → ClO + O_2

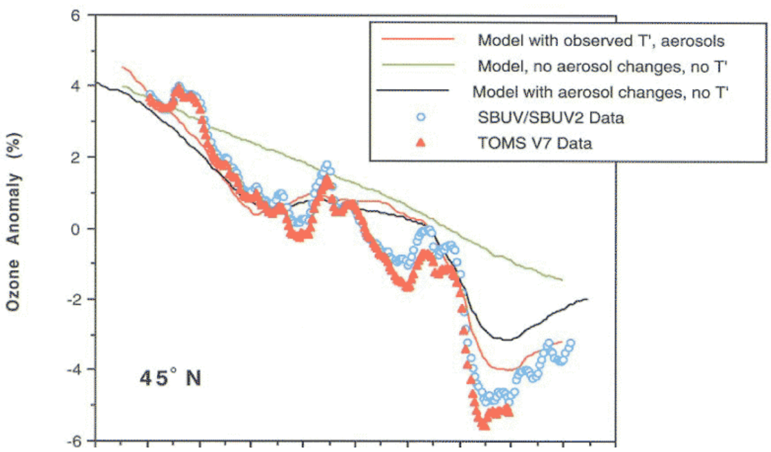
Net: 2 O_3 → 3 O_2

Polar Stratospheric Clouds

ClO_x/Cl_y & NO_x/NO_y

Aerosols (Pinatubo)

Lower Stratosphere Ozone Loss



What determines Ozone

- Source budgets: determines radicals budget
  (emissions, transport, decay)
- Family budgets: determines radicals budget
  (source decay, transport)
- Repartitioning: determines radicals budget
  (chemistry, particle budgets)
- Particle budgets: determines radicals budget
  (emission, microphysics, volcanic eruptions,...)

(temperature, illumination,...)

Nitrogen Source, N_2O

Data Assimilation: MIPAS constrained N_2O budget

↓

Chemical forecast (same dynamical fields)

↓

Assessment of the evolution of N_2O

Case study: Sept., 2002 Reminder non-operational MIPAS

Assimilation system

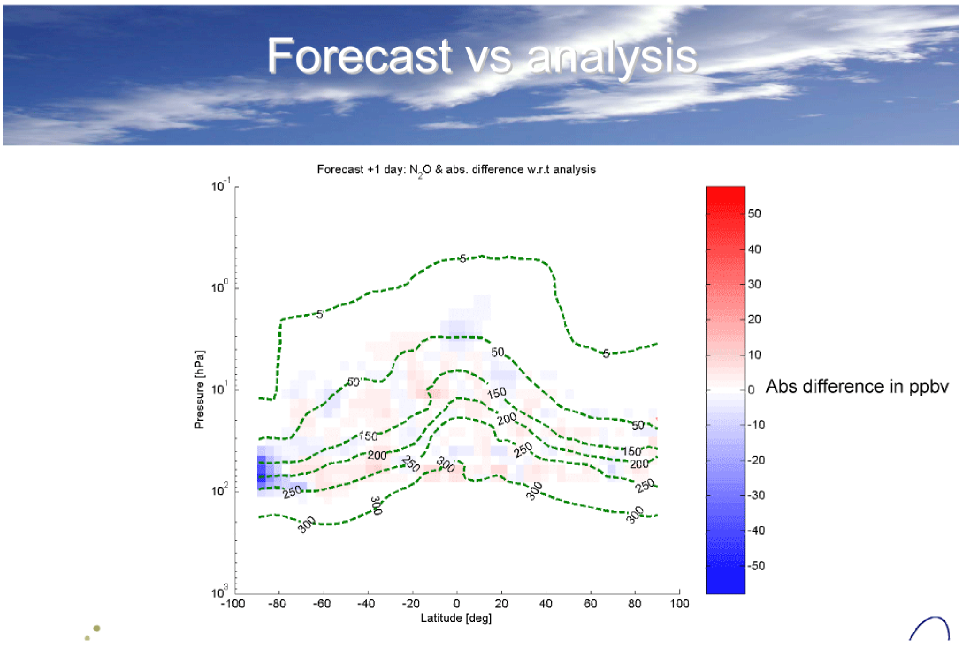
4D-var: (Talagrand & Courtier, 1987)

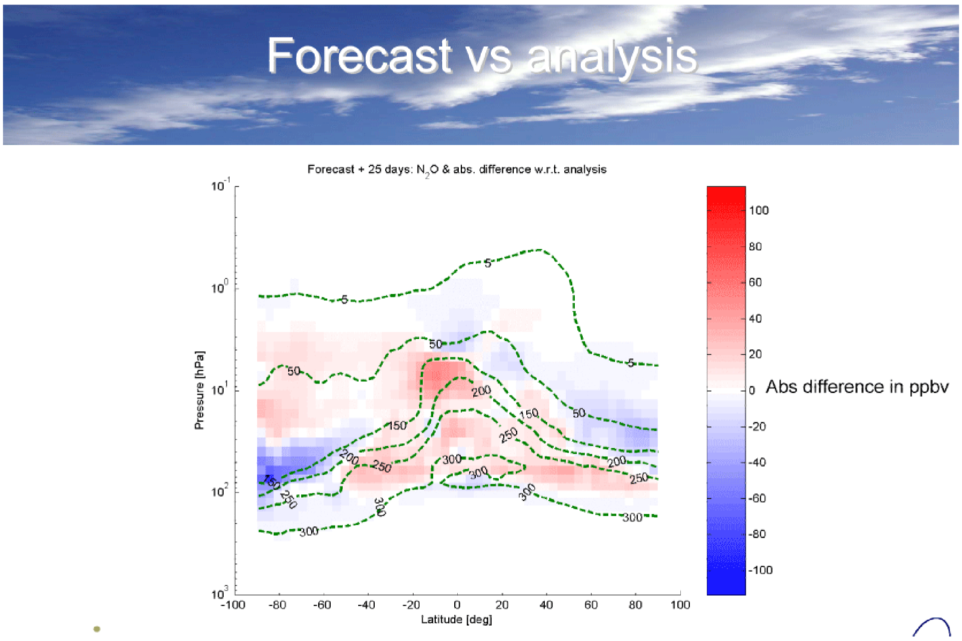
Model grid: horizontal: 3°.75 x 3°.75 (96 x 49 pts)
vertical: 37 pressure levels, surface → 0.1 hPa
(subset of ECMWF hybrid levels, keeping stratospheric levs)

Chemistry: 57 chemical species

PSC: 4 types of PSC particles (36 size bins)

Dynamics: Eulerian
driven by ECMWF 6h analyses
advection by Lin & Rood (1996) with 30' time step





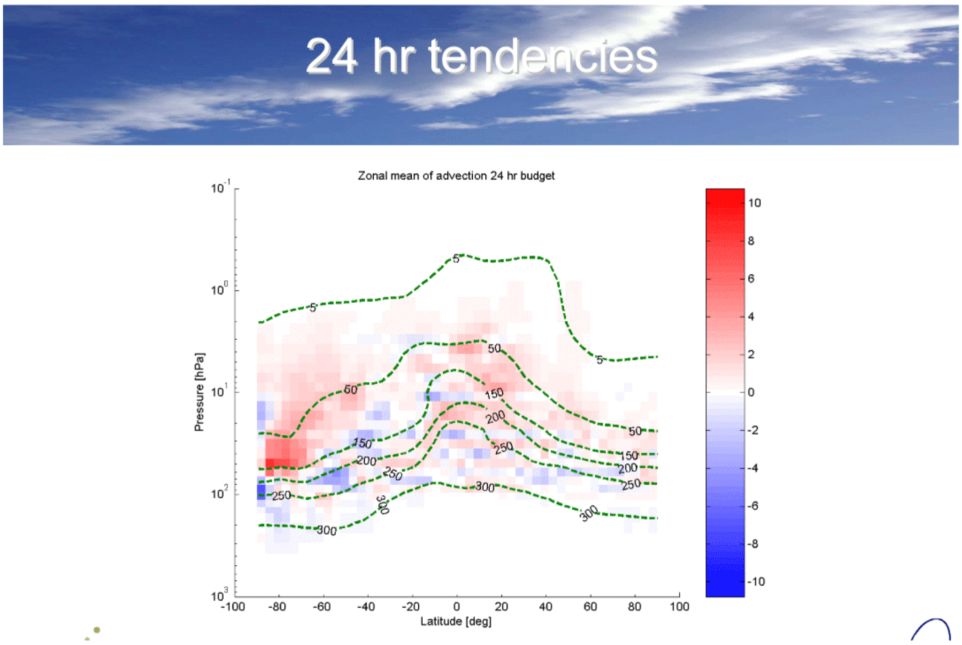
Cause of difference

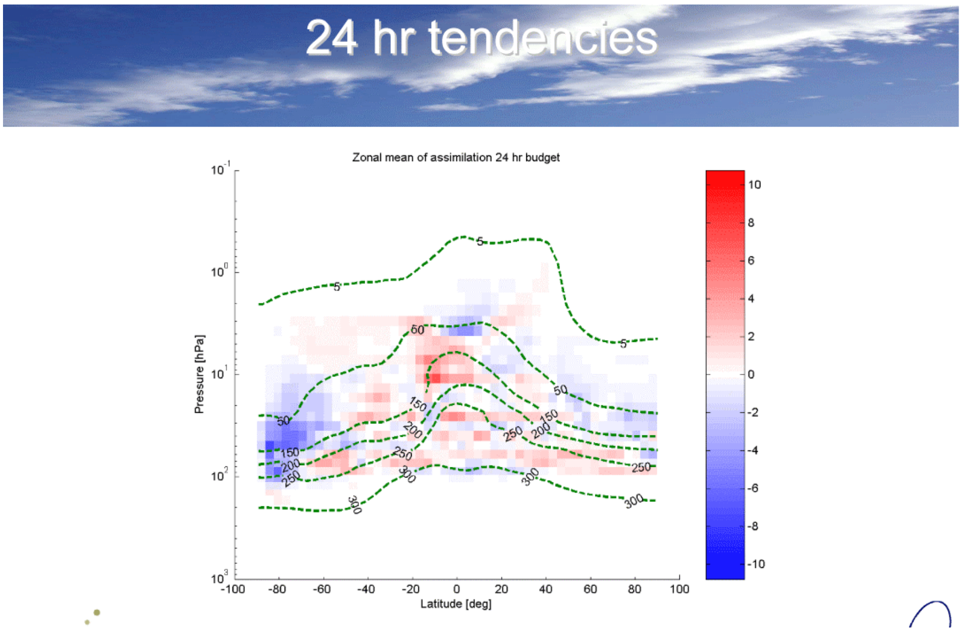
↓ (4D – VAR, history)

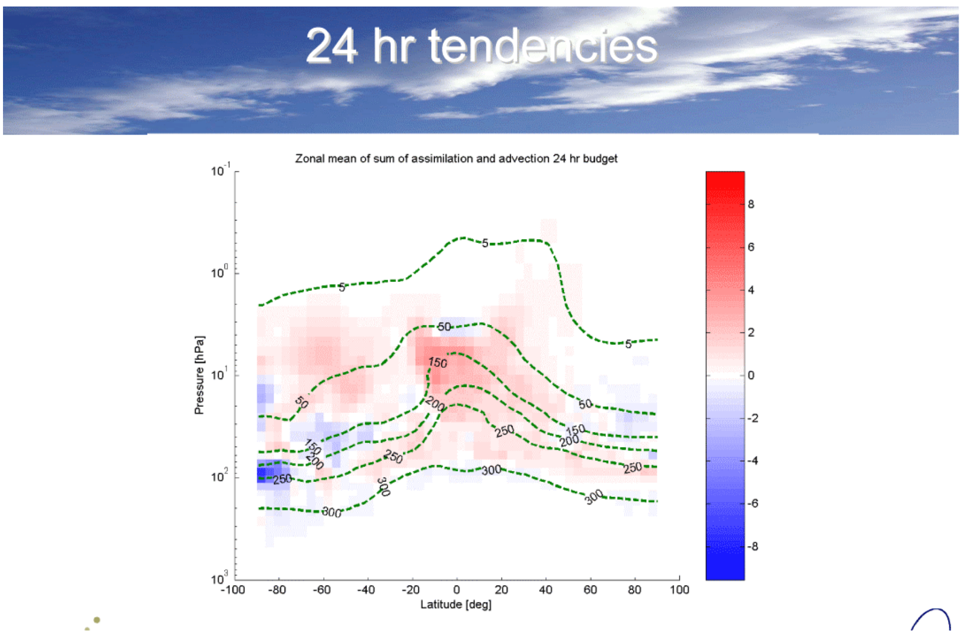
Local advection / chemistry tendency
(24 hr integrated tendency)

↓

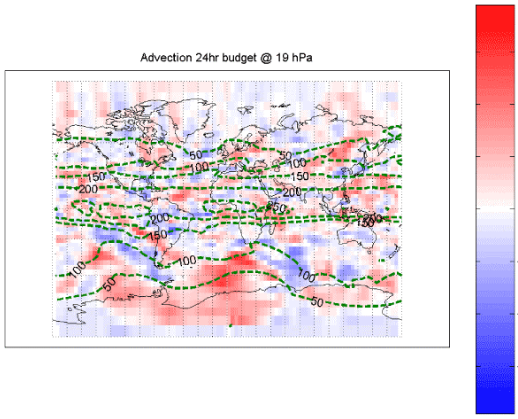
Comparison with local assimilation "tendency": (analysis – first guess)$_{t0}$
September mean


24 hr tendencies
Zonal mean of advection 24 hr budget
Pressure [hPa]
Latitude [deg]

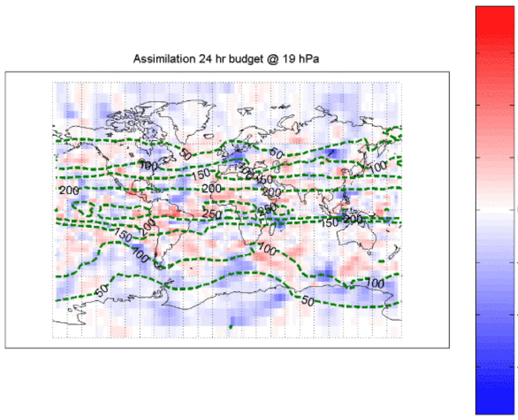

24 hr tendencies
Zonal mean of assimilation 24 hr budget
Pressure [hPa]
Latitude [deg]


24 hr tendencies
Zonal mean of sum of assimilation and advection 24 hr budget
Pressure [hPa]
Latitude [deg]

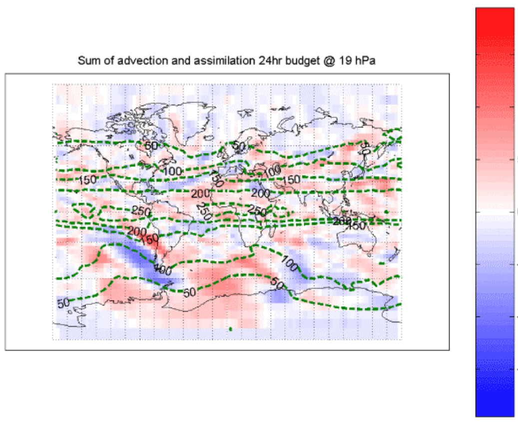
24 hr tendencies

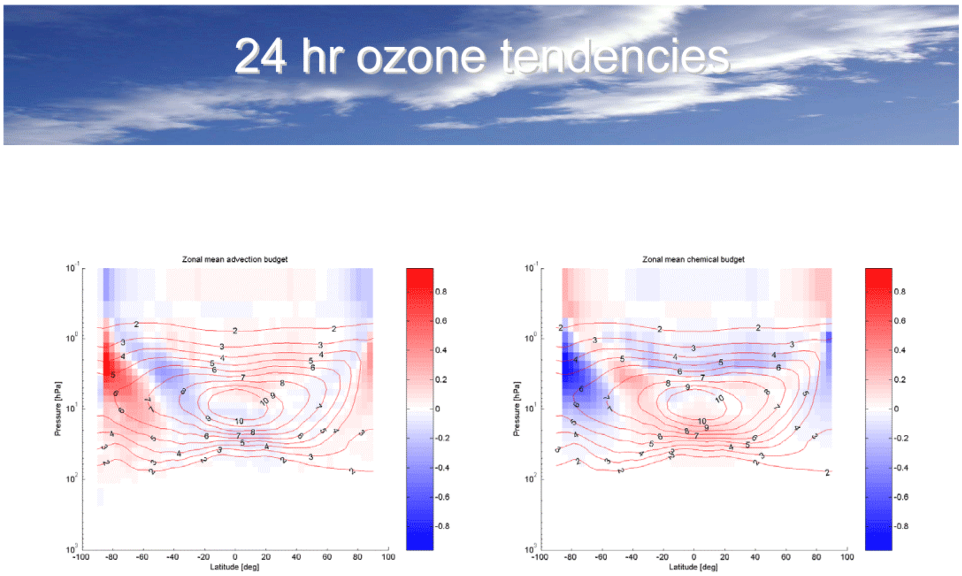


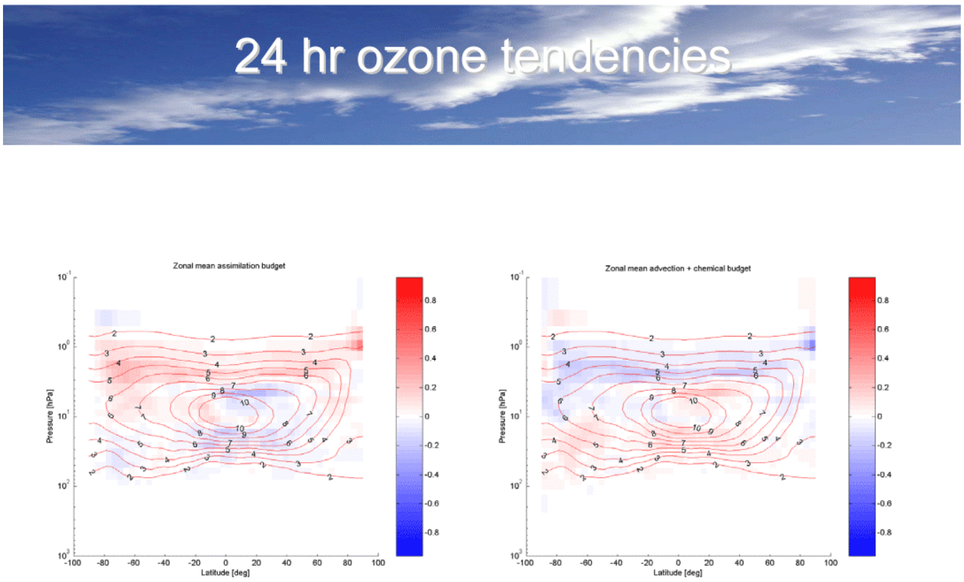
24 hr tendencies

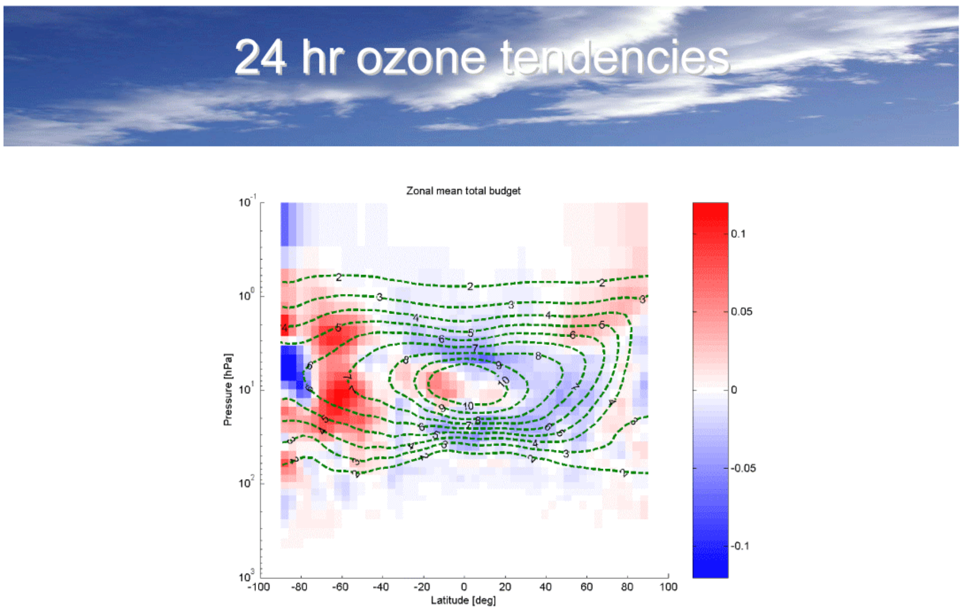


24 hr tendencies




24 hr ozone tendencies
Zonal mean advection budget
Zonal mean chemical budget
Pressure [hPa]
Latitude [deg]


24 hr ozone tendencies
Zonal mean assimilation budget
Zonal mean advection + chemical budget
Pressure [hPa]
Latitude [deg]


24 hr ozone tendencies
Zonal mean total budget
Pressure [hPa]
Latitude [deg]